MAR 20 1978

Item 830-H-15

NAS1.60:1144

NASA Technical Paper 1144

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NASA

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# Optical and Holographic Storage Properties of Fe-, Cu-, and Mg-Doped Lithium Niobate

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**Scientific and Technical
Information Office**

1978

## SUMMARY

Several samples of iron-, copper-, and magnesium-doped lithium niobate were tested to determine their optical and holographic storage properties which would be applicable to an optical data storage system and an integrated optics data preprocessor which makes use of holographic storage techniques. The parameters of interest are the diffraction efficiency, write power, write time, erase time, erase energy, and write sensitivity. Results of these parameters are presented.

It was found that iron-doped lithium niobate samples yielded the best results in all parameters except for a few percent higher diffraction efficiency in copper-doped samples. The magnesium-doped samples were extremely insensitive and are not recommended for use in holographic optical data storage and processing systems.

## INTRODUCTION

The use of satellites to record Earth resources, meteorological, and environmental pollution data requires the management of large quantities of data ($10^{10}$ to $10^{12}$ bits of data per orbit). In order to have greater spatial and spectral resolution, the data rates will consequently increase. It will be necessary not only to process these large amounts of data, but also to store vast amounts of data for subsequent transmission to the Earth.

Optical parallel processing (ref. 1) and holographic data storage (refs. 2 and 3) are attractive approaches to these data management problems. However, optical preprocessors and optical storage systems require the use of a material which can be readily recorded, read, and erased optically. Preliminary investigations have identified lithium niobate as a likely material for use in these systems. This paper is concerned with the laboratory measurement of various parameters of lithium niobate critical to the successful operation of optical preprocessors and storage systems.

## SYMBOLS

| | |
|---|---|
| $E_o$ | incident energy density, $J/cm^2$ |
| N | reciprocal of grating spacing, lines/mm |
| n | index of refraction |
| $\Delta n$ | change in index of refraction |
| S | write sensitivity, $cm^2/J$ |
| V | visibility |

| | |
|---|---|
| $\eta$ | diffraction efficiency |
| $\eta_{max}$ | maximum diffraction efficiency |
| $\theta$ | angle from normal to sample surface, deg |
| $\Lambda$ | grating spacing, mm/line |
| $\lambda$ | wavelength, nm |
| $\Phi$ | read beam angle of incidence from normal, deg |

Notations:

| | |
|---|---|
| Cu | copper |
| Fe | iron |
| $LiNbO_3$ | lithium niobate |
| Mg | magnesium |

## THEORY

An ideal holographic storage material will experience a large rapid change in its diffraction efficiency $\eta$ under illumination by laser light. It will also retain this change in diffraction efficiency until it is deliberately erased.

The diffraction efficiency $\eta$ in a material depends on the modulation of the index of refraction $n$. The change in the index of refraction $\Delta n$ of an electro-optic material depends on the electro-optic coefficient of the material and the intensity of the field pattern stored. The internal electric field pattern has a magnitude that is dependent on the number of electron traps and the energy depth of the traps. It is believed that this field pattern generated by the migration of photo-excited electrons from traps will result in optical storage in the form of thick-phase holograms in single-crystal electro-optic materials. (See ref. 4.)

The lithium niobate material used in the study reported in this paper has large electro-optic coefficients with localized concentrations of traps having electrons that can be excited by light of appropriate wavelength. A certain percentage of these traps must be empty or the trap must have the potential of trapping an additional electron. (See fig. 1(a).) In the process of writing a hologram, a light interference pattern exists in the material, and electrons are excited from the traps to the conduction band at rates dependent on the intensity of the light at a given point. This condition produces an inhomogeneous concentration of free carriers that either diffuse thermally, drift under applied or internal electric fields, or become retrapped primarily in regions of low light intensity. These processes result in a net space-charge pattern

that is positive in regions of high intensity and negative in regions of low intensity (fig. 1(b)). A field is generated by the space charge that modulates the index of refraction producing a phase hologram. The amount of incident light energy per unit area required to produce a desired diffraction efficiency is a measure of the sensitivity of the material. The sensitivity determines the exposure time required to store a hologram at a certain power density.

When the image stored in the hologram is erased, the electrons are uniformly reexcited from the traps and rearrange themselves evenly throughout the bulk of the material. This redistribution of electrons removes the field and erases the hologram. Readout of the hologram is accomplished by illuminating the hologram with a laser beam of longer wavelength than was used to write. A major problem related to the utilization of ferroelectric materials as an optical storage medium is some optical erasure of the hologram during readout; however, thermal fixing can reduce the amount of erasure.

Impurity doping of ferroelectric materials introduces a new set of traps and changes material properties in many cases. If the dopant concentration is increased, the optical absorption and sensitivity will increase if additional free electrons are produced. Also, bulk properties such as transport properties may be modified and can affect the storage time and erasure process. The transition metals are good choices for dopants since they enter the ferroelectric $LiNbO_3$ lattice substitutionally and have potential to give up additional electrons under visible light excitation.

## EXPERIMENTAL DESCRIPTION

The holographic recording behavior of doped $LiNbO_3$ was examined by writing a diffraction grating in several $LiNbO_3$ samples with a variable wavelength argon laser and simultaneously determining the grating diffraction efficiency with a continuous wave He-Ne laser ($\lambda$ = 632.8 nm). The argon laser has two high energy lines at 514.5 nm (green) and 488 nm (blue-green). However, illumination of the $LiNbO_3$ for diffraction efficiency determination at these wavelengths causes erasure or destruction of the gratings; therefore, the 632.8-nm line from the He-Ne laser was necessary since it will not erase the hologram. Gratings were written in iron-, copper-, and magnesium-doped samples at various argon laser beam energy levels at 488 nm. An iron-doped sample was tested at various recording beam wavelengths. All the samples were doped at the same concentration level.

The optical arrangement used to write diffraction gratings and determine diffraction efficiencies in doped $LiNbO_3$ is shown in figure 2. A photograph of the setup is shown in figure 3. Both the argon and helium-neon laser beams in figure 2 are vertically polarized and the c-axis of the material is positioned perpendicular to both the beam polarization and beam path. Beam splitter 1 is used to attenuate the write beam intensity. The object and reference beams have equal path lengths and beam splitter 2 is set to provide beams of equal intensity. Both the zero-order and first-order diffracted beam intensities are monitored during the write process by optical power meters and a strip chart recorder.

For both the tests where the write beam intensity and wavelength are varied, the object and reference beams impinge on the material surface at an angle of 15° from the normal to the sample surface $\theta$. Information about the sinusoidal grating produced in the sample at this angle is found from the formula:

$$N = \frac{2 \sin \theta}{\lambda}$$

where $N$ is the reciprocal of the grating spacing and has the dimensions of lines/mm.

For a write wavelength of 488 nm used in the variable write intensity test, $N$ for the resulting grating is 1060 lines/mm. The read beam angle of incidence from the normal $\Phi$ is established from the formula:

$$\Phi = \sin^{-1} \frac{\lambda_r}{2\Lambda}$$

where

$\lambda_r$ read beam wavelength, 632.8 nm

$\Lambda$ grating spacing, 1/N

A read angle of 19.6° is necessary to read out the 1060 lines/mm diffraction grating.

Mirrors 4 and 5 of figure 2 are used to make fine adjustments in $\Phi$ because of the critical nature of the read beam angle of incidence described by the formula:

$$\Delta\Phi_{1/2} = \frac{\Lambda}{2d}$$

where

$\Delta\Phi_{1/2}$ read beam angular half-power bandwidth

$d$ grating thickness

This formula (ref. 5) is applicable to thick, lossless transmission holograms. For the 1060 lines/mm grating which is 1 mm thick, $\Delta\Phi_{1/2}$ is 1.62 minutes.

To begin the experiment, all three beams were alined so that they were incident on the same spot on the material surface. With the write beam power adjusted to about 40 mW, the write wavelength adjusted to 488 nm, and the read beam intensity set to 10 mW; a diffraction grating was written in the sample. Then, using mirrors 4 and 5, $\Phi$ was adjusted to obtain the maximum intensity

of the diffracted or first-order beam. This first-order beam intensity is related to the diffraction efficiency $\eta$ by the formula:

$$\eta = \frac{\text{First-order intensity}}{\text{Zero-order intensity} + \text{First-order intensity}}$$

After acquiring the maximum possible $\eta$, the grating was erased by using the 514.5-nm wavelength line of the argon laser. The erasure involved blocking the object beam path and allowing the full intensity (about 270 mW) of the 514.5-nm wavelength reference beam to be incident upon the material. Then, a new grating was written by unblocking the object beam and returning the write beam intensity and wavelength to 40 mW and 488 nm, respectively. This new grating was written so that further adjustments in $\Phi$ could be made, and thus insure that the maximum diffraction efficiency was being recorded. There were no noticeable effects from read-write-erase cycling.

With all the beams properly alined and the correct angles established, the variable write energy test was performed. During the test, the wavelength of the write beam was left at 488 nm and diffraction gratings were written in the doped $LiNbO_3$ samples by using incident powers ranging from 10 to 100 mW. After recording information from each grating about the undiffracted or zero-order beam and the first-order diffracted beam, the readout angle was readjusted slightly to see whether the diffracted beam intensity could be increased. Repetitions of the test at the individual power levels were made to establish predictable data trends. Each grating was once again erased by the high intensity 514.5-nm wavelength line of the argon laser.

In the variable wavelength test, an iron-doped sample was tested with write wavelengths of 476.5 nm, 496.5 nm, and 501.7 nm to compare with the results obtained at 488 nm. The write incident power for all the runs was 30 mW, which was found to be a moderate power level. Since a change in the write beam wavelength is related to a change in the read beam angle, a great deal of readout angle adjustment and fine tuning was necessary for each different wavelength test. The selection of wavelengths for the variable wavelength experiment depended upon their availability from the argon laser, their maximum power levels, and the capability of the experimental setup to accommodate the resulting readout angle.

## RESULTS AND DISCUSSION

The results of the experiments are presented in figures 4 to 7 and in table I for various characteristics of lithium niobate ($LiNbO_3$) doped with chemical impurities. Figure 4 is a plot of the diffraction efficiency against write time for Fe-, Cu-, and Mg-doped $LiNbO_3$. The write wavelength was 488 nm and the power density was 0.231 $W/cm^2$. The Mg-doped samples had an extremely low diffraction efficiency $\eta$ and are eliminated from further discussion. The Fe-doped $LiNbO_3$ sample reached maximum $\eta$ in about 50 seconds, whereas the Cu-doped sample had a slightly higher $\eta$, but required approximately a factor of 10 longer write time to reach maximum $\eta$.

Figure 5 is a plot of write power density against time to obtain $\eta$ = 10 percent for the best samples of Fe- and Cu-doped $LiNbO_3$. This type of curve is meaningful because the point on the curve where $\eta$ = 10 percent is on the linear portion of the curve for $\eta$ against write time. It also is at a point where thermal effects, etc., due to the write process have not produced any fluctuations. For a power density below approximately 0.3 $W/cm^2$, the Fe-doped samples require much less time for writing the diffraction grating than do the Cu-doped samples. For a higher power density (>0.3 $W/cm^2$), the write time for Fe-doped samples is slightly less than that for Cu-doped samples.

Figure 6 is a plot of diffraction efficiency as a function of the elapsed erasure time for the diffraction grating in $LiNbO_3$. The power density is 2.15 $W/cm^2$ and the erase beam wavelength is 514.5 nm. The $\eta$ for the samples was 60 percent before the erase procedure was initiated. The Fe-doped samples were rapidly erased and reached 50 percent erasure in about 10 seconds, and essentially total erasure in about 40 seconds. The Cu-doped samples require more than twice the time to erase than the Fe-doped samples do. Total erasure of the Cu-doped samples was not fully achieved until after several minutes of erasure. This condition could pose a problem in the continuous formation and erasure of holograms in the material since a buildup of background noise could cause errors in data formation and readout.

Figure 7 is a plot of maximum $\eta$ in Fe-doped $LiNbO_3$ as a function of write time where various wavelengths of an argon ion laser have been used to write the diffraction gratings. The write power density was kept at a constant 0.231 $W/cm^2$. The write $\lambda$ of 496.5 nm required about 40 seconds to produce the maximum $\eta$, whereas write wavelengths of 476.5, 488, and 501.7 nm required about 55 seconds to produce the maximum $\eta$ with very little difference between them. This result shows some wavelength dependence in the Fe-doped samples. Of the wavelengths examined, a wavelength of 496.5 nm is the most effective wavelength for writing a hologram. However, the 488-nm line of the argon ion laser is the most powerful and was chosen as the write wavelength for the tests previously discussed.

## CONCLUDING REMARKS

Several samples of iron-, copper-, and magnesium-doped lithium niobate were tested to determine their optical and holographic storage properties which would be applicable to an optical data storage system and an integrated optics data preprocessor which makes use of holographic storage techniques. Both Fe- and Cu-doped samples had high diffraction efficiencies $\eta$. The Mg-doped $LiNbO_3$ exhibited extremely low $\eta$ (25 percent) and extremely large write and erase times. Even though $\eta$ for Cu-doped $LiNbO_3$ is slightly higher than for Fe-doped, the Cu-doped $LiNbO_3$ required significantly longer write and erase times. The Cu-doped samples showed a write time which was longer by a factor of four and an erase time which was longer by a factor of three. Most important was the need of about five times the write power density for Cu-doped samples to reach an $\eta$ of 10 percent. The write sensitivity of Cu-doped samples was more than an order of magnitude lower than Fe-doped samples.

Iron-doped $LiNbO_3$ was better than the copper doped in every area except the maximum diffraction efficiency attained where only a few percent difference was noted. From the improvements noted in the doping of $LiNbO_3$ in these tests, it is believed that further research with doped $LiNbO_3$ will produce a ferroelectric material that will be useful in holographic-type memories and integrated optics processing devices which employ holography.

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January 26, 1978

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TABLE I.- RESULTS OF HOLOGRAPHICALLY STORED DIFFRACTION GRATINGS IN DOPED LITHIUM NIOBATE

| Type of dopant | Typical diffraction efficiency, $\eta$, percent | Maximum diffraction efficiency, $\eta_{max}$, percent (a) | Write power density, $W/cm^2$ (b) | Material absorption, percent (c) | Write time for $\eta$ = 10 percent, sec | Write energy density for $\eta$ = 10 percent, $J/cm^2$ (d) | Erase time for 1/e, sec (e) | Erase energy density for 1/e, $J/cm^2$ (d) | Write sensitivity, S, for $\eta$ = 10 percent, $cm^2/J$ (f) |
|---|---|---|---|---|---|---|---|---|---|
| Fe | 60 | 67 | 0.231 | 63 | 7 | 1.0 | 10 | 14 | 0.20 |
| Cu | 65 | 70 | .231 | 75 | 28 | 5.0 | 32 | 48 | .049 |
| Mg | 25 | 28 | .231 | 26 | 1120 | 67 | 1530 | 859 | .0012 |

[a]For 30 mW write power.

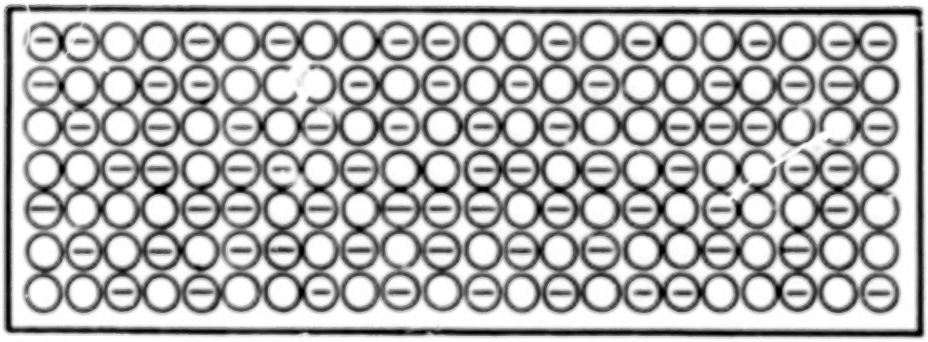
[b]At incident surface.

[c]$\lambda$ = 488 nm.

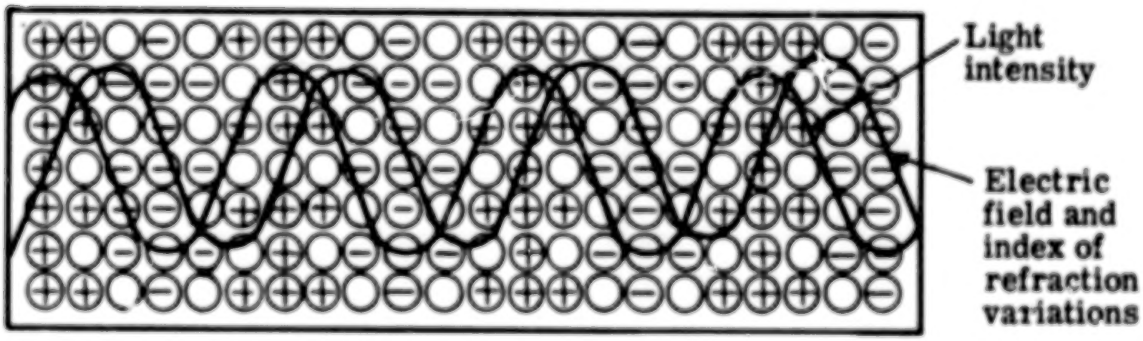
[d]Write and erase energy includes only that absorbed by the samples (reflected and transmitted energy is not included).

[e]1/e is 36.8 percent of $\eta_{max}$ where e is the exponential.

[f]$S = \frac{\eta^{1/2}}{E_o V}$ where V is the visibility (1.0 for beams of equal intensity) and $E_o$ is the incident energy density.



(a) Trap distribution before exposure to light.




(b) Charge distribution after exposure to light (ref. 3).

Figure 1.- Electro-optic storage medium.

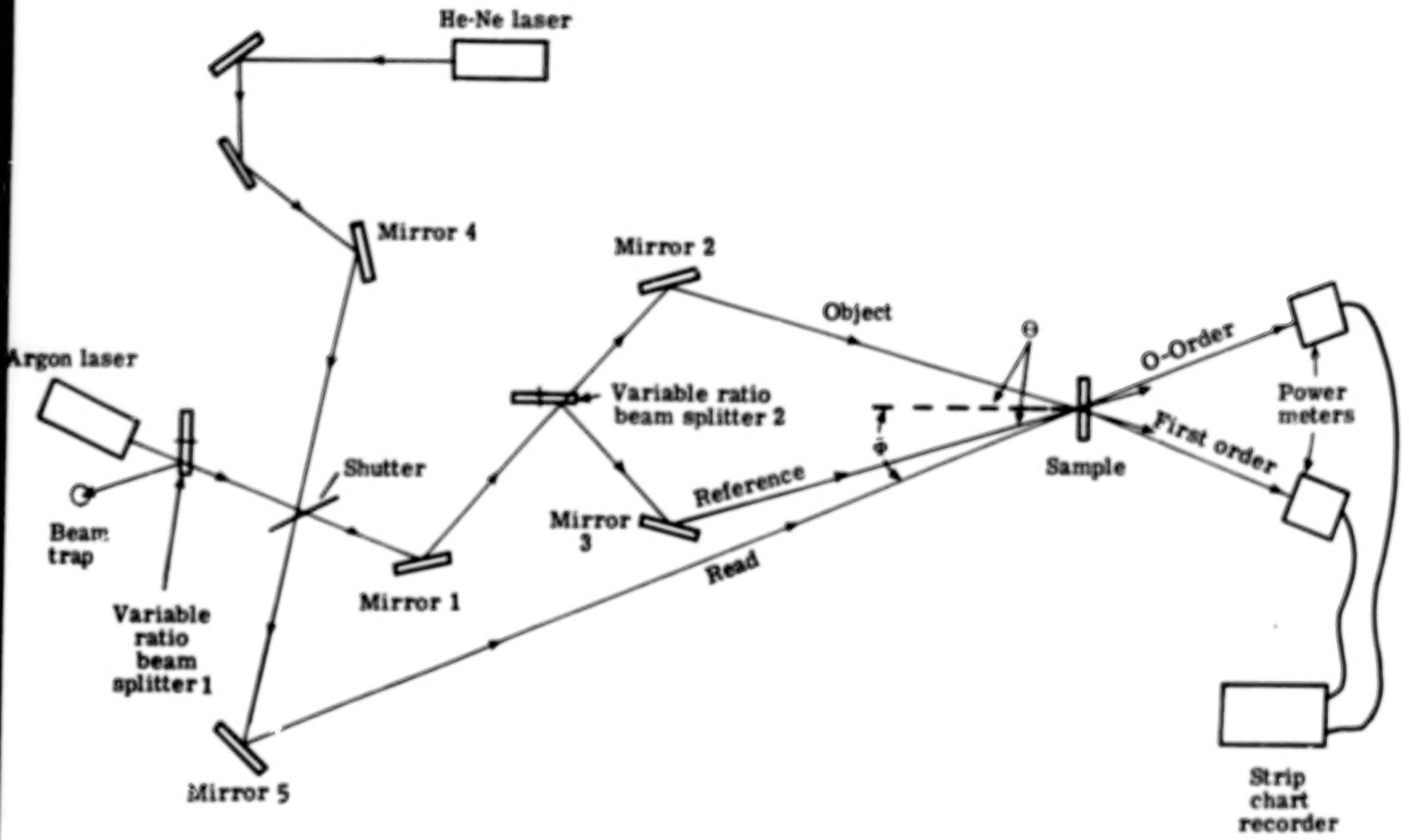



Figure 2.- Optical arrangement.

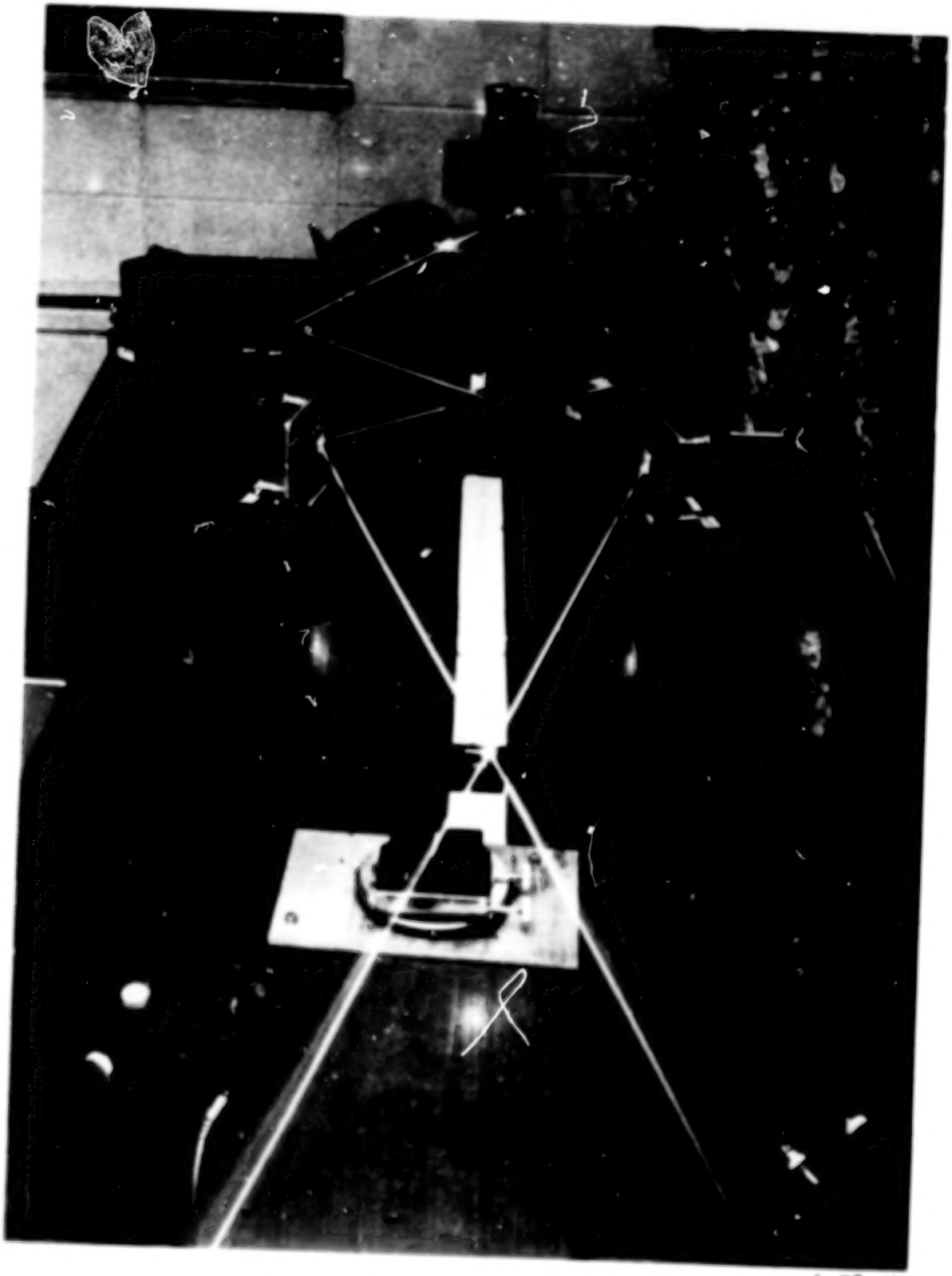


Figure 3.- Experimental setup.

L-78-14

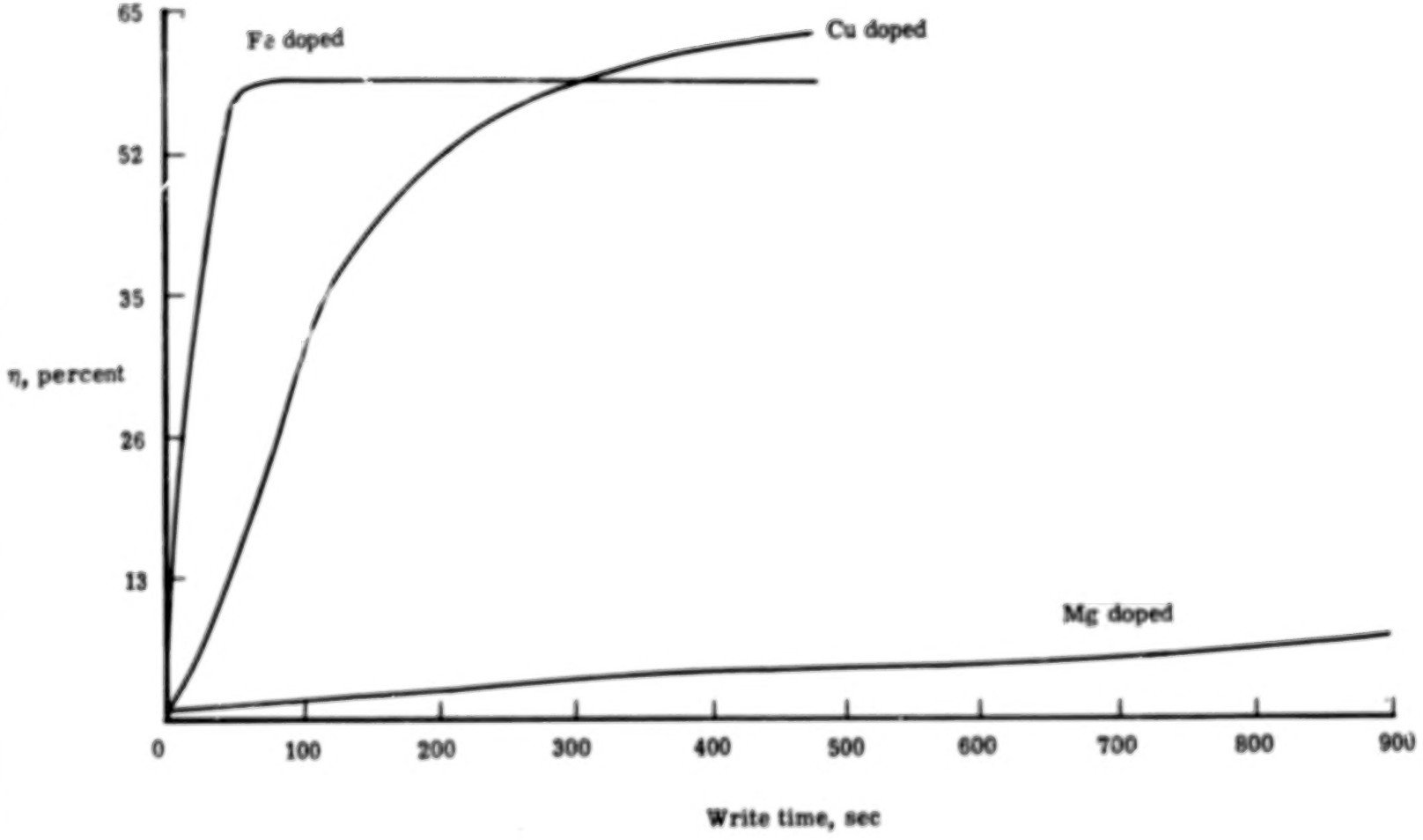



Figure 4.- Write characteristics of doped $LiNbO_3$. Power density = 0.231 $W/cm^2$; $\lambda$ = 488 nm.

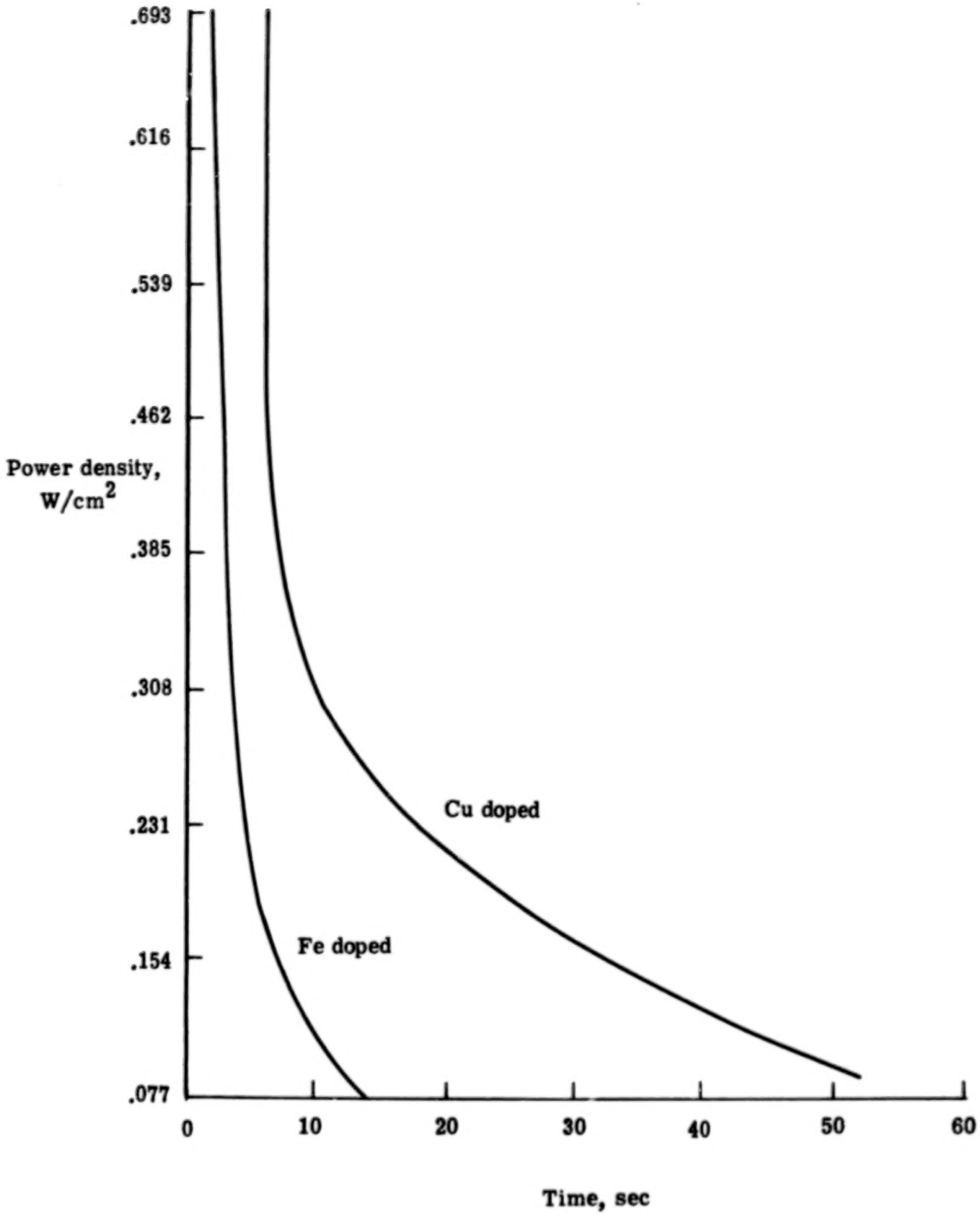



Figure 5.- Write power density plotted against time for 10 percent $\eta$ in doped $LiNbO_3$. Write $\lambda$ = 488 nm.

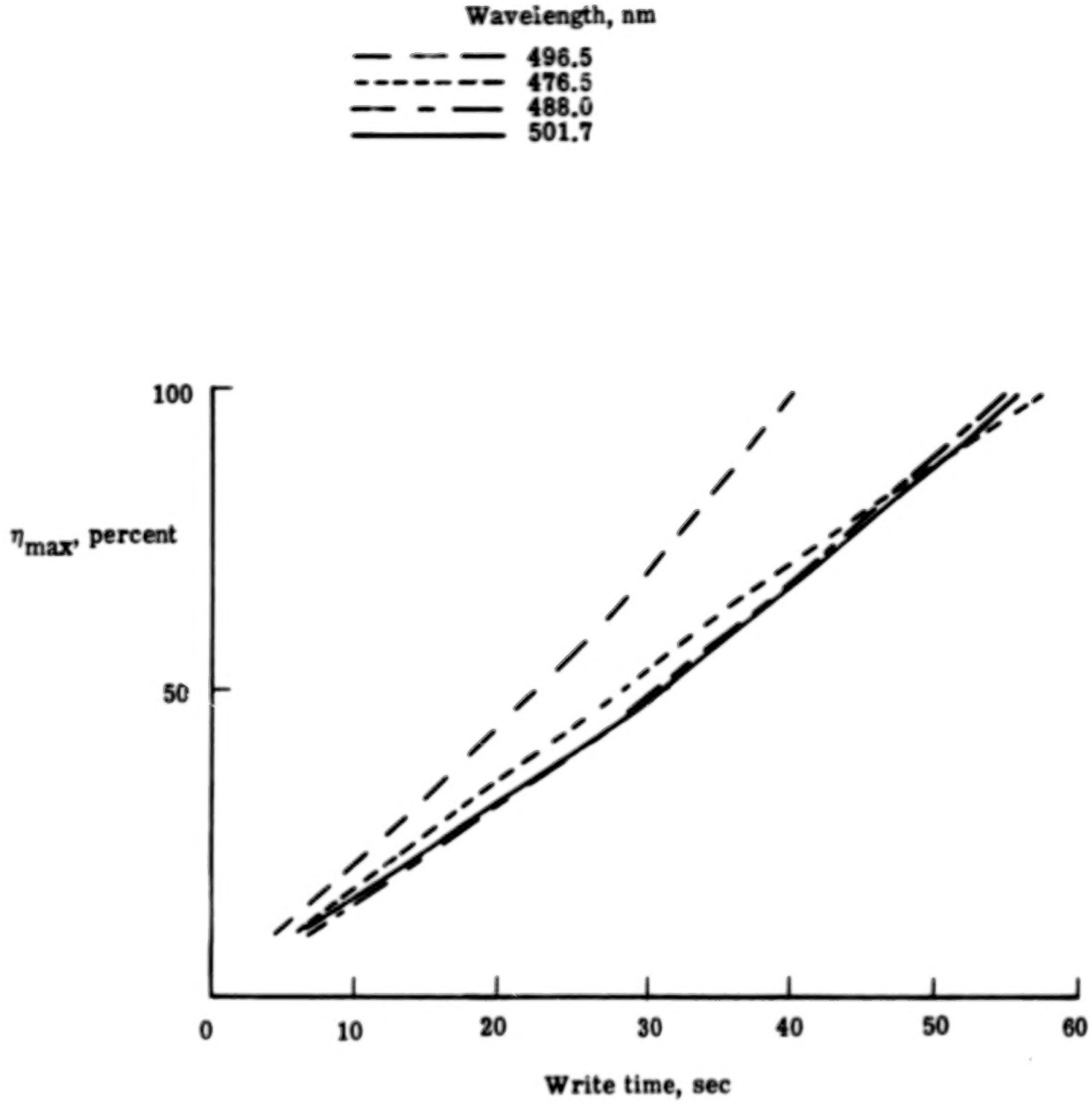



Figure 7.- Results of variable write wavelengths in Fe-doped $LiNbO_3$. Power density = 0.231 $W/cm^2$.

| 1. Report No. | 2. Government Accession No. | 3. Recipient's Catalog No. |
|---|---|---|
| NASA TP-1144 | | |

| 4. Title and Subtitle | 5. Report Date |
|---|---|
| OPTICAL AND HOLOGRAPHIC STORAGE PROPERTIES OF Fe-, Cu-, AND Mg-DOPED LITHIUM NIOBATE | March 1978 |
| | 6. Performing Organization Code |
| 7. Author(s) | 8. Performing Organization Report No. |
| Marvin E. Beatty III and Barry D. Meredith | L-11964 |
| | 10. Work Unit No. 506-18-21-01 |
| 9. Performing Organization Name and Address | 11. Contract or Grant No. |
| NASA Langley Research Center<br>Hampton, VA 23665 | |
| | 13. Type of Report and Period Covered Technical Paper |
| 12. Sponsoring Agency Name and Address | 14. Sponsoring Agency Code |
| National Aeronautics and Space Administration<br>Washington, DC 20546 | |

15. Supplementary Notes

16. Abstract

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| 17. Key Words (Suggested by Author(s)) | 18. Distribution Statement |
|---|---|
| Holographic storage<br>Lithium niobate<br>Optical data storage systems<br>Integrated optics | Unclassified - Unlimited<br><br>Subject Category 74 |

| 19. Security Classif. (of this report) | 20. Security Classif. (of this page) | 21. No. of Pages | 22. Price* |
|---|---|---|---|
| Unclassified | Unclassified | 15 | $4.00 |

* For sale by the National Technical Information Service, Springfield, Virginia 22161

NASA-Langley, 1978